(2)

REPORT DOCUMENTATION PAG-

AD-A239 059

1. AGENCY USE ONLY (Leave blank)

2. REPORT DATE

3. REPORT TYPE AND DATES COVERED
Final Report 01 Dec 88-31 Oct90

4. TITLE AND SUBTITLE
SCIENTIFIC IMAGING SYSTEM

5. FUNDING NUMBERS
AFOSR-89-0152

6. AUTHOR(S)
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7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
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8. PERFORMING ORGANIZATION REPORT NUMBER
AFOSR-TR- 91 0655

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
AFOSR/NE
BLDG 410
BOLLING AFB DC 20332-6448

10. SPONSORING/MONITORING AGENCY REPORT NUMBER
2917/A3

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT
UNLIMITED

12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 words)

EQUIPMENT WAS PURCHASED ON THIS GRANT

14. SUBJECT TERMS

15. NUMBER OF PAGES

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

NSN 7540-01-280-5500

Standard Form 298 (Rev. 2-89)
Prescribed by ANSI Std. Z39-18

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June 7, 1991

Dr. Alan Rosenstein
Air Force Office of Scientific Research
Bolling Air Force Base
Building 410
Washington, DC 20332

SUB: Final Report for the "Scientific Imaging System" Grant #AFOSR-89-0152

Dear Alan,

Acquisition of all the necessary components for the scientific imaging system based on absorption and fluorescence spectroscopy for the measurement of concentration and nucleation rate during non-equilibrium synthesis by laser is almost complete. The list of equipment for the assembly of this unique diagnostic system is enclosed as Attachment A. Major components include a 20 W Argon-Ion laser and a Ring-Dye laser with various associated electronics. Equipment which has already been delivered and is currently working are marked in Attachment A. The remaining few are in the process of acquisition, awaiting the evaluation of the core laser scanning and detection system. This phased purchase will allow us to specify the remaining electronics to match the performance of core-laser scanning system.

The diagnostic technique using this system will involve performing absorption and fluorescence spectroscopy on the plasma produced by a laser beam impinging upon the metallic surface. The diagnostic spectroscopy will use a tunable dye laser (Coherent 899-28) to probe a particular transition originating in the ground state of niobium and aluminum. The absorption cross-section at the wavelength of interest [(5252 A°(Nb) and 3961A°(A1)] may be calculated using the tabulated values for the oscillator strengths of the various transitions between hyperfine levels which contribute to absorption at this wavelength. Using this value for the cross-section and the well-known Beer-Lambert law for the intensity of a beam, I(L), after passage through a region of length, L, containing an absorbing medium with absorption cross-section, σ,

$$I(L) = I_0 e^{-NL\sigma},$$

the number density, N, within the absorbing volume may be determined, assuming the initial intensity of the beam is known. Thus the number density may be obtained.

Implementation of the above technique is straightforward in a situation where the absorbing medium uniformly fills up some volume, i.e. a vapor in equilibrium. The plasma volume in the laser ablation experiments does not, however, fulfill this condition. Rather it is expected that the plasma will occupy a roughly dumbell-shaped region centered about the laser beam which initiates the plasma. The density of metal atoms in this volume most likely will vary spatially. The dye

laser probe beam will therefore pass through regions of nonuniform density. This may be corrected for by assuming cylindrical symmetry for the plasma volume and considering that the total absorption measured by the beam will be an Abel transform of the density distribution. The transform may be inverted by a computer program to give the true density distribution. The length of the region through which the beam propagates (that is, the region over which the plasma volume extends along the line of the laser beam) may be determined by imaging the fluorescence (or perhaps even the total optical emission) from the plasma volume. The resonance fluorescence is perhaps best for this, since it will indicate only regions where the metal atoms are being excited. A final problem stems from the temperature dependence of the absorption cross-section. An independent method of determining the temperature in the plasma volume must be employed to establish the correct value for the cross-section.

The absorption spectroscopy technique suffers from the fact that it is not particularly sensitive. It requires the detection of small changes in the laser beam intensity against the rather large amplitude-noise that is characteristic of laser output. The minimum density observable with this method is on the order of 10^9 atoms/cm^3. Densities as low as 100 atoms/cm^3 for sodium vapor have been obtained by Fairbank, Hänsch, and Schawlow [1] using a resonance fluorescence technique. That is they observe the fluorescence component which is at the same wavelength as the exciting dye laser. This is not the optimum case, since scattered laser light from the cell cannot be distinguished easily from the true fluorescence. The lack of any easily detectable intermediate fluorescence determined the choice. A number of optical and single-averaging techniques were employed to minimize this problem. The 525.2 nm transition in Nb and 396.1 nm transition in Al were recommended by Fairbanks, Hänsch, and Schawlow [1] in their paper as being particularly suitable for this type of measurement.

The most novel aspect of the Fairbanks-Schawlow method was that it provided an independent calibration of the absolute number density without a dependence on tabulated pressure versus temperature curves. This was managed by only observing a portion of the fluorescence region with length, L_{obs}. The total resonance fluorescence in the region is simply proportional to the total integrated absorption in the region. That is, any absorbed light comes out as fluorescence. The fluorescence intensity, I_F, is then,

$$I_F = I_i - I_T$$

or

$$I_F/I_i = 1 - e^{-N\sigma L_{obs}}$$

where I_i is the intensity of the beam at the beginning of the observation region. This, however, from the Beer-Lambert law is just,

$$I_i/I_0 = 1 - e^{-N\sigma L_{arm}}$$

where L_{arm} is the length of the region prior to the observation region. Thus, I_F may be expressed,

$$I_F/I_0 = e^{-N\sigma L_{arm}} (1 - e^{-N\sigma L_{obs}})$$

A plot of this function reveals that I_F will go through a maximum at a unique density, dependent only on the cross-section and the lengths of the two regions. Thus, by varying the vapor density (pressure) to find the value at which fluorescence peaks, assigning to that condition the calculated

peak density, and referring all subsequent measurements to that density value, absolute density measurements of the species of interest in the vapor phase may be obtained.

Applying this refinement to the spatially nonuniform plasma volume created by the laser presents several difficulties. Yet, even if the absolute calibration should prove impractical, useful relative density measurements may be extracted from this technique.

The main use for this laser spectroscopy is to measure the nucleation rates of the alloying process. For this method, a pulsed and CW beam will be employed. The dye laser will once more be tuned to be resonant with a transition of one of the component atoms. The time decay of fluorescence (or absorption) on this transition should reflect the depletion of the (free) component species from the plasma volume. While variants of this method are regularly employed to monitor chemical processes, to our knowledge it has never been done to investigate the combination of metal vapors.

One of the interesting study will be to influence the nucleation of certain compounds by pumping the participating ion into different energy levels using laser induced fluorescence. If successful, this will enable us to actively participate in the synthesis process.

Doppler-free saturation spectroscopy [2] developed at Stanford has a potential for extreme contrast improvement and line width reduction for optically thick atomic samples. This transmission spectroscopy technique has the possibilities in characterizing the plasma during interaction of metal targets with high intensity laser beam where a plasma sample may be optically thick. We would like to explore the possibilities of applying this technique to understand atomic species under an intense field during laser materials interaction. Simplification of complicated spectrum is another objective worth pursuing. Several methods have already been developed by Professor Schawlow and his co-workers [2-4] and applied for Na_2.

To the best of our knowledge, this is probably the first such attempt to experimentally explore the basic science of nucleation during alloying. Let me take this opportunity to thank you for providing us with the necessary support to attempt such a challenging project.

With best wishes and regards,

Yours sincerely,

J. Mazumder
Professor of Mechanical Engineering

JM/ajs
Enclosures: Attachment A
References
xc: Grants & Contracts Office
Jeff Oberg, M&IE Business Office

References:

1. Fairbanks, Jr., W., T. W. Tänsch, and A. L. Schawlow, "Absolute Measurement of Very Low Sodium-vapor Densities using Laser Resonance Fluorescence," *J. of the Optical Society of America*, Vol. 65, No. 2, pp. 199-204, February 1975.

2. Svanberg, G-Y. Yan, T. P. Duffey, W. M. Du, T. W. Hänsch, and A. L. Schawlow, "Saturation Spectroscopy for Optically Thick Atomic Samples," *J. of the Optical Society of America B*, Vol. 4, pp. 462-468, April 1987.

3. Carlson, N. W., K. M. Jones, G. P. Morgan, A. L. Schawlow, A. J. Taylor, H-R., Xia, and G-Y. Yan, "Selective Spectrum Simplification by Laser Level Labeling," *Proc. of the IV Conference on Laser Spectroscopy*, Jasper, Canada, June 29-July 3, 1981.

4. Kaminsky, M., R. T. Hawkins, F. V. Kowalski, and A. L. Schawlow, *Physical Review Letters*, Vol. 36, p. 671, 1976.

Attachment A

Revised Equipment List for Scientific Imaging of Laser Processing

	Qty.	Total Price
√(1) Coherent Innova 200-20 Ar Ion Laser	1	$ 61,047
√(2) Coherent 899-29 AutoScan Dye Laser (cw) w/Optics for DCM dye	1	81,000
√(3) Additional Mirror Sets for Dye laser		
(i) Coumarin 6	1	3,008
(ii) Ex 392E	1	3,008
√(4) Lambda Physik LPD302E Pulsed Dye Laser	1	35,500
(5) IBM PS/2-30 w/Metrabyte Data Acquisition Board	1	4,400
(6) Ethernet Board for (4)	1	300
√(7) Newport KNS-48-18 Optical Table (TMC instead of Newport)	1	4,953
√(8) Newport XLAH Table Legs (Again, TMC)	3	2,300
√(9) Newport XL-C Casters for Table Legs	3	710
√(10) Newport ATS-8 Shelf System	1	1,600
√(11) Coherent 216 Spectrum Analyzer		
(a) Mirrors $1,570		
(b) Tube 1,890		
(c) Detector 280		
(d) Mount 600		
(e) Beam Spl. 280		
(f) Controller 1.465		
SUBTOTAL	1	6.085
(12) Additional Mirror Set for Coherent 216 Spectrum Analyzer 350-450 nm Operation	1	2,000
√(13) Coherent FieldMaster Power Meter Console	1	895
√(14) Coherent LM-10 Detector Head for (12)	1	930
√(15) LM-200 Sensitive Range Head for (12)	1	595
(16) Stanford Research Sys SR-S10 Lock-in Amp	1	2,990
√(17) Isomet 1206C AO Modulator	1	930
√(18) Isomet D320 Deflector Driver	1	930
√(19) Hamamatsu R928 Photomultiplier Tube	1	365
√(20) Hamamatsu E371-16 Base for (18)	1	50
√(21) Hamamatsu HV Power Supply for (18)	1	850
√(22) EG&G DT-25 Photodiodes	4	130
√(23) EG&G FND-100 Photodiodes	4	200

√(24) Clean Rooms Int. Flow Hood for Dye Laser	1	600
√(25) NesLab Dye Chiller Module	1	1,495
√(26) Tektronix 2224 Dig/Analog Scope w/GPIB	1	4,795
√(27) BK Precision Function Generator	1	600
(28) Hewlett-Packard 6206B Power Supply	1	950
√(29) Hewlett-Packard 623B Triple Power Supply	1	800
√(30) Klinger Scientific SL25.4 Mirror Mount	1	405
√(31) Newport 461XYZ Translation Stage	1	695
√(32) Newport 461Z Translation Stage	1	310
√(33) Klinger MR6 XYZ Stage (Cat. No. 3339063)	1	700
√(34) Klinger MRL8.25 Z Trans. Stage (Cat. No. 338052)	1	485
√(35) Newport 481 Rotary Stage	1	321
√(36) Melles-Griot 07BLJOOl Lab Jack	1	495
√(37) Newport lOZ40DMS Steering Mirror	1	122
√(38) CVI Laser F06520-100 Interference Filter	1	95
(39) Amperex XX1410 Image Intensifier/CCD Array w/Camera Module	1	4,300
(40) Video Cassette Recorder	1	300
(41) EG&G 46 Elem. Parallel Output Photodiode Array	1	310
√(42) Miscellaneous (Optical Magnetic Mounts, Posts, Mirror Mounts, Lens Mounts, Mirrors, Lenses, Cables, Electronics, etc.) [Much of this has been purchased]	1	5,809
√(43) Sun Sparc Station 330 for Data Processing	1	29,190
√(44) Hitachi Color Printer for Producing Hard Copies of Processed Data (Purchased Seiko Inst 5500 Series instead)	1	5,000
(45) Processing Chamber (w/Vacuum Pump, Optical Ports and Necessary Control System)	1	10.175
SUBTOTAL		$282,728
Coherent has offered to accept partial payment in the sum of $97,214 for the dye laser/argon laser combination. The balance will be carried over to be paid in the future.		- 44,833
Cost sharing from Research Board for the Sun Spark Station		- 24,000
Cost sharing from the Department		- 10,000
TOTAL		**$203,895**